

TUNNELING CURRENTS AND THE E - k RELATION*

Stephen Kurtin,[†] T. C. McGill,[‡] and C. A. Mead
California Institute of Technology, Pasadena, California 91109
(Received 18 May 1970)

The energy-momentum dispersion relation within the forbidden gap of a single-crystal insulator (in this case, GaSe) has been accurately determined using a simple physical model to describe tunneling currents in appropriate thin-film structures. This dispersion relation, calculated from experimental current-voltage data, is shown to be intrinsic to GaSe and capable of quantitatively predicting tunneling currents. The work reported here represents the first quantitative calculation of tunneling currents in metal-insulator-metal structures with all parameters relevant to the experiment independently determined.

The band structure of covalent solids¹ includes an energy range in which quantum mechanically allowed states do not exist.² The nature of this forbidden gap,³ although important to a complete understanding of the electronic behavior of solids,⁴ is relatively unexplored because the forbidden gap is inherently inaccessible to electrons. Only in extremely thin structures ($<100 \text{ \AA}$) can a significant number of electrons tunnel through the forbidden region, and in so interacting yield information about the energy-momentum dispersion relation over this energy range.⁵

Previous work on the determination of $k(E)$ from tunneling currents has employed current-voltage data obtained either from Schottky-barrier diodes⁶⁻⁸ formed on crystalline material or from thin-film structures incorporating amorphous insulating films.⁹⁻¹¹ In the former case, both the thickness of the depletion layer and the energy of the final state depend on the applied bias. The inability to separate physically these two variables complicates the otherwise straightforward analysis of tunneling. Attempts at confirming a $k(E)$ relation derived from Schottky-barrier measurements are usually performed by varying the doping level. A change in carrier concentration affects the depletion-layer shape and offers a quasi-independent check of $k(E)$. Experiments must be confined to highly doped specimens to assure narrow depletion layers and hence maintain tunneling as the primary mechanism of current flow. Meaningful measurements can be made only on materials where excellent control of doping level can be obtained.

In principle, the thin-film structure is better suited for the study of dispersion relations via analysis of tunneling currents since the thickness of the forbidden region is independent of applied bias. The ability to check directly the exponential dependence of tunneling currents on insulator thickness removes much potential ambiguity con-

cerning the physics underlying current flow.⁹ However, the usual experimental methods of fabricating thin films have dictated the incorporation of amorphous films within thin-film structures. These films are inherently ill-defined and often spatially and chemically nonuniform. In such cases, it is not possible to characterize the structure exactly and results are often difficult to interpret. Beyond these experimental problems, there is also the fundamental theoretical question of exactly what property of an amorphous material is actually measured by the tunneling experiment—perhaps some average over the attenuation constant in various crystallographic directions.

Thus, a thin-film structure consisting of a well-defined single-crystal material sandwiched between two metal electrodes would be ideal for the study of tunneling currents. The energy-band representation of a structure fabricated to meet these criteria is shown in the inset to Fig. 1. Our approach to the fabrication of well-defined structures is to use as the insulating film a material with an anisotropic, layered crystal structure which facilitates peeling a single-crystal specimen to the required thinness ($<100 \text{ \AA}$). The layer compound gallium selenide (GaSe) was chosen as the thin-film insulator in this investigation because it has been sufficiently studied previously^{12,13} that many parameters of a thin-film structure are known *a priori* from direct experiments on the parent bulk GaSe crystal. From such data the shape of the potential barrier in a metal-GaSe-metal structure is anticipated to be, to excellent approximation, trapezoidal as represented in the inset to Fig. 1. Possible deviations from this simple shape arising from space-charge or image-force effects are negligible.¹²

Since the basic idea behind electron tunneling is an exponentially damped wave function, a WKB integral is usually used to express the de-

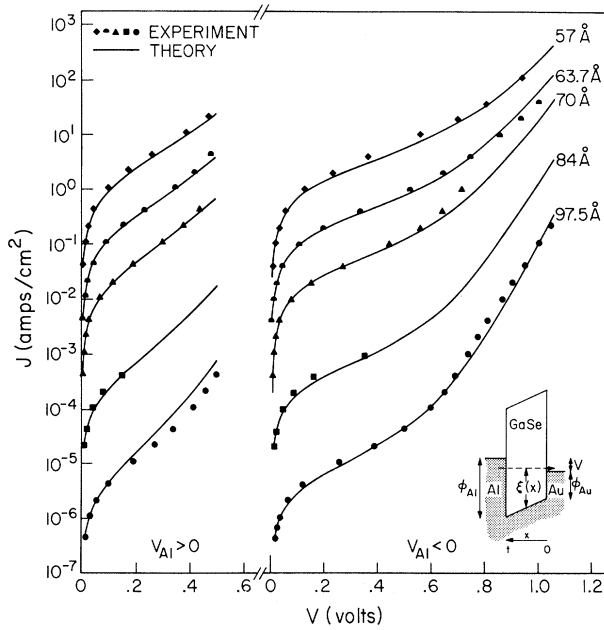


FIG. 1. Current density J as a function of applied voltage V for several Al-GaSe-Au structures. Data for both bias directions are shown as the solid symbols. Theoretical curves are calculated from the measured $E-k^2$ dispersion relation for GaSe (Fig. 2) via the WKB approximation [Eq. (1)]. The thickness indicated for each structure is that determined from structure capacitance. These experimentally determined thickness values are used in all calculations. The inset shows a band-diagram representation of a typical Al-GaSe-Au structure. This band diagram is known to be accurate from prior measurements performed on bulk specimens of GaSe. $\xi(x)$ is the energy as a function of position of a tunneling electron (measured from the valence band).

pendence of the tunneling probability [and hence $J(V)$] on $k(E)$. For a metal-insulator-metal structure with a trapezoidal barrier, the electric field within the insulating region is uniform and hence an appropriate WKB formulation (in the zero-temperature limit) may be expressed as

$$J(V) = \left(\frac{e}{2\pi\hbar} \right) \frac{\varphi_2 - \varphi_1 - eV}{t \int_{\varphi_1 + E}^{\varphi_2 + E - eV} d\xi / k(\xi)} \int_0^{eV} dE \times \exp \left\{ - \frac{2t}{\varphi_2 - \varphi_1 - eV} \int_{\varphi_1 + E}^{\varphi_2 - eV + E} k(\xi) d\xi \right\}, \quad (1)$$

where φ_1 and φ_2 are the metal-GaSe barrier energies, and V is the applied bias. This equation is, of course, only valid for that range of applied bias for which direct interelectrode tunneling is possible: $V < \varphi_1$, $V < \varphi_2$.

Given the availability of experimental $J(V)$ data

over the entire direct-tunneling bias range,^{14,15} it is conceptually straightforward to invert Eq. (1) and obtain the $k(E)$ dispersion relation for the thin-film insulator incorporated within the structure under study. Computationally, the development of a suitable technique is required. There are approximation techniques based on series expansion of Eq. (1), or on the dominant exponential dependence of tunneling current on insulator thickness, which can be profitably applied to this problem. These techniques usually require some assumption about the energy distribution of tunneling electrons which, although well satisfied in many cases, cannot be known to be valid short of self-consistency tests applicable after the $E-k$ relation has been determined. A more general approach is the direct numerical inversion of Eq. (1). An inversion technique¹⁶ which converges nicely to a unique $k(E)$ has been employed in the analysis of the experimental data reported here.

Data obtained on a series of Al-GaSe-Au structures are shown by the solid symbols in Fig. 1. These data were obtained at 77°K. The apparent thickness of each structure was determined directly from capacitance measurements and used in all calculations. Preliminary analysis¹² of the data confirms the dominance of direct tunneling as a mechanism of current flow and hence indicates that detailed numerical analysis of the data is worthwhile. Therefore, these $J(V)$ data (for each insulator thickness) were assumed to correspond to the mechanism of Eq. (1), and inverted to obtain the experimentally determined $k(E)$. Each value of insulator thickness yielded a substantially identical E vs k^2 curve; these curves were averaged to obtain a best approximation $E-k^2$ dispersion relation for GaSe. The resulting experimentally determined energy-momentum dispersion relation of GaSe is shown in Fig. 2. This relation is parabolic near the valence band, as expected, but k has extraordinarily little dependence on E toward mid-gap. This lack of substantial dependence of k on E results in a distribution of tunneling electrons which covers a wide energy range, and hence the usual approximation techniques for obtaining the $E-k^2$ relation from $J(V)$ would have been inadequate for the analysis of tunneling currents in GaSe.

A good check of the accuracy and self-consistency of this experiment is to calculate, using no adjustable parameters,¹⁷ the tunneling currents predicted by Eq. (1) and the measured $E-k$ curve. The theoretical $J(V)$ curves of Fig. 1 have been

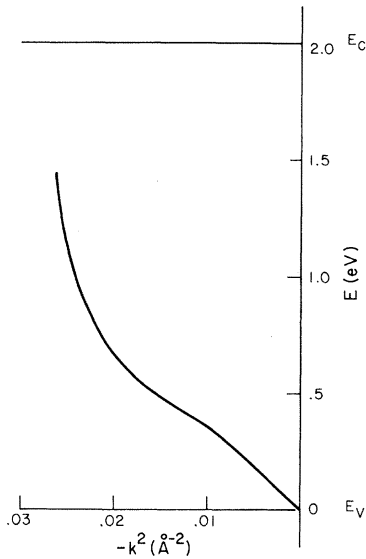


FIG. 2. The measured $E-k^2$ dispersion relation in the forbidden gap of GaSe. Negative values of k^2 indicate the square of the imaginary part of k ; i.e., the electronic attenuation coefficient. This curve is calculated using the known potential barrier shape (see inset to Fig. 1) of Al-GaSe-Au structures and the experimental $J(V)$ characteristics of these structures. Near the valence band the $E-k^2$ relation may be represented by the effective-mass approximation with $m^* = 0.07$. As shown, that portion of the $E-k^2$ curve near the conduction band was inaccessible to our measurements.

calculated on this basis. Agreement between theory and experiment is seen to be excellent for all voltages and thicknesses. This agreement is noteworthy because any one $J(V)$ curve contains enough information to specify $k(E)$ completely. The ability to calculate $J(V, t)$ quantitatively over the entire relevant range of both voltage and insulator thickness indicates that the theoretical model is an excellent representation of the physical situation. For the thinner specimens, currents observed at 300°K were nearly equal to those observed at 77°K, as anticipated. We conclude that direct electrode-electrode tunneling through trapezoidal barriers is the mechanism of current flow and that the simple WKB model is adequate to describe quantitatively tunneling in Al-GaSe-Au structures.

To demonstrate that the $E-k^2$ relation determined from the above analysis is indeed an accurate, intrinsic, and fundamental property of GaSe (rather than a property dependent upon the particular electrodes used) further experimentation is required. By choosing an electrode metal other than aluminum, copper for example, the

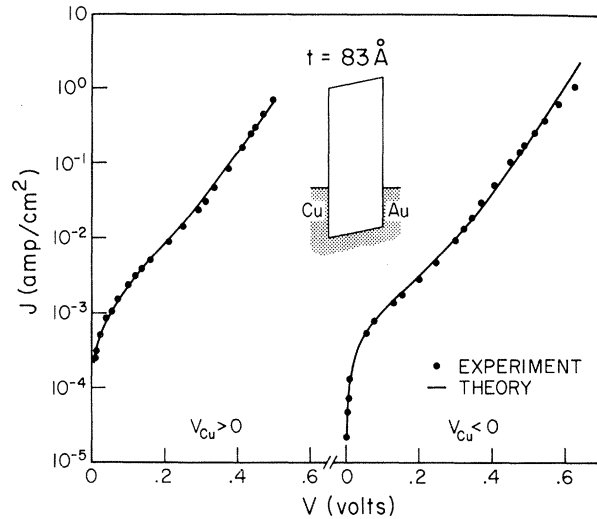


FIG. 3. Current density J as a function of applied voltage V for a Cu-GaSe-Au structure. Experimental data are shown as the solid circles. The theoretical curve was calculated from the $E-k^2$ relation of Fig. 2 via the WKB approximation [see Eq. (1)]. Quantitative agreement between theory and experiment confirms that the measured $E-k^2$ relation is an accurately determined intrinsic property of GaSe.

GaSe-metal barrier energy may be altered and a structure with a band diagram quantitatively different from that of an Al-GaSe-Au structure obtained. Electrons tunneling in a Cu-GaSe-Au structure encounter a range of k for every applied bias which is distinct from that encountered in the Al-GaSe-Au structure (compare the inset of Fig. 1 with that of Fig. 3; $\phi_{Al} = 1.08$ eV, $\phi_{Cu} = 0.68$ eV). Therefore, if the $E-k^2$ relation of Fig. 2 predicts tunneling currents in Cu-GaSe-Au structures it must be both an intrinsic property of GaSe and accurately determined.

Figure 3 presents data obtained on a Cu-GaSe-Au structure containing an 83-Å film of single-crystal GaSe. The theoretical curve, calculated directly¹⁷ from Eq. (1) using the $E-k^2$ relation of Fig. 2 and the known properties of GaSe, also appears in the figure. Theory and experiment are seen to be in excellent agreement thus confirming that the energy-momentum dispersion relation of Fig. 2 is an intrinsic property of GaSe.

In summary, the ability to fabricate well-defined, single-crystal, thin-film structures has provided experimental data which are in quantitative agreement with a simple model of current flow by direct interelectrode tunneling. The energy-momentum dispersion relation within the forbidden gap of a solid has been accurately

determined by the analysis of tunneling currents. This technique allows the forbidden range of energies in the complex band structure of a solid to be probed directly.

*Work supported in part by the U. S. Office of Naval Research.

†Hertz Foundation Doctoral Fellow.

‡Howard Hughes Doctoral Fellow.

¹S. Kurtin, T. C. McGill, and C. A. Mead, *Phys. Rev. Lett.* **22**, 1433 (1969).

²See, for example: H. Ehrenreich, H. R. Philipp, and J. C. Phillips, *Phys. Rev. Lett.* **8**, 59 (1962); D. Brust, J. C. Phillips, and F. Bassani, *Phys. Rev. Lett.* **9**, 94 (1962); and others.

³F. Herman, R. L. Kortum, C. D. Kuglin, J. P. Van Dyke, and S. Skillman, in *Methods in Computational Physics*, edited by B. Alder, S. Fernbach, and M. Rotenberg (Academic, New York, 1968), Vol. 8, p. 193.

⁴See, for example: E. O. Kane, *J. Phys. Chem. Solids* **12**, 181 (1959); V. Heine, *Proc. Phys. Soc., London* **81**, 300 (1963); R. O. Jones, *Proc. Phys. Soc., London* **89**, 443 (1966); E. I. Blount, in *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1962), Vol. 13, p. 362.

⁵C. A. Mead, *Tunneling Phenomena in Solids* (Plenum, New York, 1969), p. 127.

⁶F. A. Padovani and R. Stratton, *Phys. Rev. Lett.* **16**, 1202 (1966).

⁷J. W. Conley and G. D. Mahon, *Phys. Rev.* **161**, 681

(1967).

⁸G. H. Parker and C. A. Mead, *Phys. Rev. Lett.* **21**, 605 (1968), and *Appl. Phys. Lett.* **14**, 21 (1969), and *Phys. Rev.* **184**, 780 (1969).

⁹G. Lewicki and C. A. Mead, *Phys. Rev. Lett.* **16**, 939 (1966).

¹⁰D. Meyerhofer and S. A. Ochs, *J. Appl. Phys.* **9**, 2535 (1963).

¹¹J. C. Fisher and I. Giaever, *J. Appl. Phys.* **32**, 172 (1961).

¹²See, for example, T. C. McGill, S. Kurtin, and C. A. Mead, to be published, and references therein.

¹³S. Kurtin and C. A. Mead, *J. Phys. Chem. Solids* **29**, 1865 (1968).

¹⁴For a discussion of techniques for the identification of current flow mechanism see, for example, Ref. 9, Ref. 12, or S. Kurtin, T. C. McGill, and C. A. Mead, to be published.

¹⁵We consider here only applied biases such that tunneling, if present, occurs directly from one metallic electrode to the other.

¹⁶T. C. McGill, thesis, California Institute of Technology, 1969 (unpublished).

¹⁷In all cases the thickness of the insulating film is determined directly from the capacitance of the structure (which is effectively a parallel-plate capacitor). All other parameters of the structure are known from previous measurements performed on bulk specimens. Hence, no amplitude scaling is required, and none was performed. See also S. Kurtin, T. C. McGill, and C. A. Mead, to be published.

FAR-INFRARED ABSORPTIVITY OF NORMAL LEAD*

H. Scher

Xerox Research Laboratories, Xerox Square, Rochester, New York 14603

(Received 22 June 1970)

The Holstein-Boltzmann equation has been used to calculate the far-infrared absorptivity of normal Pb in and above the phonon-frequency region. The absorptivity exhibits a threshold at $\bar{\nu} \approx 35 \text{ cm}^{-1}$ and weak structure at $\approx 70 \text{ cm}^{-1}$ related to the transverse and longitudinal peak in the phonon density of states, respectively. It increases rapidly through the phonon region, eventually saturating at the Holstein value. These results agree with the recent experiments of Joyce and Richards.

What happens when a high-frequency electromagnetic wave is incident on a pure metal at very low temperature? To the extent that the metal can be approximated by a free-electron gas there exists no mechanism for the loss of electromagnetic energy; the absorptivity is zero.¹ Some years ago, Holstein^{2,3} showed that, even at $T = 0$ and for an infinite residual resistance ratio, there exist, in actuality, two dissipative mechanisms. One is due to the presence of a surface.² The electrons acquire an oscillatory energy from the electric field in passing through the skin depth δ_f ; this energy is then converted

to heat upon diffuse collision with the surface and subsequently dissipated in the interior of the metal. The other mechanism is purely a bulk process.³ The electron simultaneously absorbs an incident photon and emits a phonon. From a quantum mechanical point of view the "free" conduction electrons are able to absorb the photon because collision with the surface or a phonon allows for the conservation of both energy and momentum. Holstein considered the high-frequency limit, i.e., $\omega \gg \omega_D$, a typical phonon frequency (e.g., for Pb, $\hbar\omega_D \approx 8.3 \text{ meV}$), where the volume absorption process is independent of